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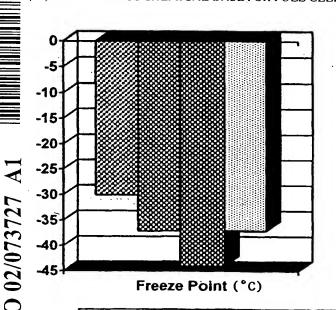
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(54) Title: A NOVEL CHEMICAL BASE FOR FUEL CELL ENGINE HEAT EXCHANGE COOLANT/ANTIFREEZE



(57) Abstract: A nontoxic fuel cell engine coolant which has an electrical resistivity of greater than 250 kOm-cm, a boiling point of greater than 90° C, a freezing point of less than -40° C, a thermal conductivity of greater than 0.4 W/m-k-, a viscosity of less than 1 mPa.s (1 cPs) at 80° C, a viscosity of less than 6 mPa.s (6 cPs) at 0° C, a heat capacity of greater than 3 kJ/kg-K, and which is compatible with current cooling system materials.

☑TC 50% ☑TC 55% ☑TC 60% ☐EG 50%

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A NOVEL CHEMICAL BASE FOR FUEL CELL ENGINE HEAT EXCHANGE COOLANT/ANTIFREEZE

Field of the Invention

This invention relates to a novel technology for use in cooling systems for fuel cell powered vehicles and/or equipment. In order to remove the heat that is generated in fuel cell systems, 1,3-propane diol is used as the chemical base for the heat exchange fluid.

Background of the Invention

It has been suggested that fuel cell technology can be used to generate electricity in sufficient volume to be applicable in the driving of electric motors for passenger vehicles, standby power generation, and other applications. A fuel cell is a device that converts chemical energy of a fuel directly into electricity and they are intrinsically more efficient than most other energy generation devices, such as internal combustion engines. In principle, a fuel cell operates somewhat like a battery. Unlike a battery, a fuel cell does not run down or require recharging. It will produce energy in the form of electricity and heat as long as fuel is The most common type of fuel cell consists of two electrodes sandwiched around an electrolyte. Oxygen passes over one electrode and hydrogen over the other, generating electricity, water, and heat.

The fact that heat is generated by the fuel cell requires the presence in the automobile or other system of a cooling system which can be similar to those used presently in internal combustion engines. Typically, such a system includes a circulating pump, plumbing that may include aluminium, brass, copper, lead-tin solder,

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stainless steel, plastic or rubber materials, and a heat exchanger (radiator) typically constructed of aluminium or copper/brass.

The heat exchange fluid (coolant) is obviously just as important in a fuel cell system as it is in internal combustion engines. Many of the requirements of a heat exchange fluid for internal combustion engines are also required for fuel cell engines. However, there are some additional requirements. For instance, fuel cell vehicles generate a direct current of 400 volts. The coolant, which flows around the aluminium components of the fuel cell, must be nonconductive to protect both the cell itself from shorting out and to prevent electrical hazard to humans operating or servicing the system.

The first fuel cell was built in 1839 by Sir William Grove, a Welsh judge and gentleman scientist. The "Grove cell" used a platinum electrode immersed in nitric acid and a zinc electrode in zinc sulphate to generate about 12 amps of current at about 1.8 volts. There were other developments in fuel cell technology over the years but serious interest in the fuel cell as a practical generator of electricity did not begin until the 1960's, when the U.S. Space Program chose fuel cell technology over nuclear power and solar energy. This technology, developed by Francis Thomas Bacon, used nickel gauze electrodes and operated under pressures as high as 2068 kPa (300 psi).

Summary of the Invention

A nontoxic fuel cell engine coolant which has an electrical resistivity of greater than 250 kOhm-cm, a boiling point of greater than 90°C, optionally, a freezing point of less than -40°C, a thermal conductivity of greater than 0.4 W/m-k, a viscosity of less than 1

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mPa.s (1 cPs) at 80°C, a viscosity of less than 6 mPa.s (6 cPs) at 0°C, a heat capacity of greater than 3 kJ/kg-K, and which is compatible with current cooling system materials. The coolant may contain from 1 to 100, preferably 40 to 85 and most preferably 55 to 85, volume percent PDO and most or all of the remaining balance is water.

Brief Description of the Drawings

Figure 1 illustrates the aqueous solution freeze point characteristics of the 1,3-propanediol and GM 6043 inhibition chemistry (EG).

Figure 2 is a plot of the freeze behaviour of aqueous 1,3-propanediol antifreeze.

Detailed Description of the Invention

As previously stated, the purpose of a fuel cell is to produce an electrical current that can be directed outside the cell to do work, such as powering an electric motor. Because of the way electricity behaves, this current returns to the fuel cell, completing an electrical circuit. The chemical reactions that produce this current are the key to how a fuel cell works. There are several kinds of fuel cells which operate somewhat differently but in general terms, hydrogen atoms enter a fuel cell at the anode where a chemical reaction strips them of their electrons. The hydrogen atoms are now "ionized" and carry a positive electrical charge. The negatively charged electrons provide the current through wires to do work.

Oxygen enters the fuel cell at the cathode and it there combines with electrons returning from the electrical circuit and hydrogen ions that have travelled through the electrolyte from the anode. In some fuel cells the oxygen picks up electrons and then travels

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through the electrolyte to the anode where it combines with hydrogen ions. This chemical reaction generates a significant amount of heat energy which must be removed from the fuel cell in order for it to continue to operate properly.

A number of objectives have been identified for coolants for fuel cell vehicles. First, since fuel cell vehicles generate a direct current of 400 volts, the coolant which flows around the aluminium components of the fuel cell must be nonconductive to protect the cell from shorting out and to prevent electrical hazards. Other physical property objectives for fuel cell coolants are set out in the table below:

Table 1

15 Electrical Resistivity >250 kOhm-cm Boiling point > 90°C Freezing point < -40°C Thermal Conductivity > 0.4 W/m-kViscosity < 1 mPa.s (1 cPs) @ 80°C 20 mPa.s (6 cPs) @ 0°C > 3 kJ/kg-KHeat Capacity Durability > 5,000 hours of operation/3 years total time Material compatibility: Compatible with current cooling 25 system materials Toxicity Classified as non-toxic for transportation

1,3-propanediol (PDO), which is manufactured by Shell Chemical Company, is generally made as described in US-A-5304691 and the art described therein. This is a process for making PDO and HPA (3-hydroxypropanal, a 3-hydroxyaldehyde). In this particular patent, PDO and HPA are made by intimately contacting an oxirane (ethylene oxide, hereinafter 'EO'), a ditertiary phosphine-modified cobalt carbonyl catalyst, a ruthenium catalyst promoter, and syngas (carbon monoxide and hydrogen) in an inert

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reaction solvent at hydroformylation reaction conditions. A PDO yield of up to 86-87 mole% is reported, using a catalyst comprising cobalt ligated with 1,2-bis(9-phosphabicyclononyl)ethane as bidentate ligand, and either triruthenium(0) dodecarbonyl or bis[ruthenium tricarbonyl dichloride] as cocatalyst. Other methods of making PDO are known.

Inhibited with the GM 6043 chemistry, the 1,3propanediol performed somewhat better than EG in modified ASTM-type tests. Figure 1 illustrates the aqueous solution freeze point characteristics of the 1,3-propanediol and GM 6043 (EG). There is a slight compromise of freeze protection as determined by the ASTM D1177 test method, but the 1,3-propanediol was soft and slushy at the reported freeze point. This could be an indication that actual protection against hard, damaging freezing is actually better, approaching the effective protection point of the EG-based product. The D1177 test was also performed with 55% and 60% 1,3-propanediol in water, and it was found that the 55% concentrated product offered protection equivalent to 50% EG, per the test method. Freeze protection continued to improve at 60% 1,3propanediol. It is felt that the antifreeze properties of the chemistry are acceptable. Indeed a 50% solution would provide adequate protection against freezing in most geographies. TC in Figure 1 is an internal designation for the PDO aqueous solutions at 50, 55, and 60 volume percent PDO.

Figure 2 shows the freeze behaviour of PDO/water

• solutions. It can be seen that formulations may be made
with freeze points significantly lower than -40°C.

It may be desirable to include an effective amount of an antifoaming composition in the antifreeze/coolant

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composition. Such components are well known. Polyglycol-type antifoaming agents can be used.

PDO coolants in fuel cell vehicles will have an electrical resistivity of greater than 250 kOhm-cm, a boiling point of greater than 90°C, usually a freezing point of less than -40°C, a thermal conductivity of greater than 0.4 W/m-k, a viscosity of less than 1 mPa.s (1 cPs) at 80°C and less than 6 mPa.s (6 cPs) at 0°C, a heat capacity of greater than 3 kJ/kg-K, a desired durability of greater than 5000 hours of operation (three years total time), material compatibility - will not corrode or erode current automotive cooling system materials, have a toxicity classified as non-toxic for transportation, and will be cost competitive with current automotive coolants.

The PDO formulations give intrinsically better protection against cavitation than EG or PG.

It is the theory that some or all of these advantages are based upon the relative chelation ability of PDO versus EO and PO. The latter are readily able to chelate the ions. The chelate with EO and PO will be a five-membered ring which is relatively easy to form. PDO cannot chelate the ions as well because it forms a six-membered ring and this is more difficult.

EXAMPLES

25 Two chemistries were used in the following experiments. These are 1,3-propane diol (anhydrous) and 1,3-propane diol (50 to 85 percent volume percent aqueous solution).

Example 1

At the beginning, it was believed that the classical corrosion and performance testing regimen as described in ASTM literature (2001 Annual Book of ASTM Standards,

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Volume 15. 05) provides an accepted method to evaluate and compare the corrosive properties of coolants to the metals customarily used in vehicle coolant systems. The new variable for fuel cells is the 400 volt (Direct Current) electric field and the issues that such a field presents to the coolant. Ionic inhibitors are disqualified. The above coolants, running in the maximum resistance state with no inhibitors, were reviewed.

It was believed that the following tests would accurately predict the above coolants' abilities to perform in a heat exchange system, in terms of corrosion protection, and physical and chemical properties. Since these new coolants had not been through this regimen of testing before, there was no experience or normal performance against which the tests could be compared for reasonableness. Therefore, each of the tests was controlled against 50 volume percent aqueous inhibited ethylene glycol.

The classical coolant development approach involves analysing the fluid for physical and chemical properties. Once the properties are established, performance objectives are determined and the prototypes evaluated. These tests may be modified to better evaluate the performance of a coolant in its intended operating environment. Examples of modifications may include variations in the pressure, temperature, electric fuel environment, and duration of the tests. The data then will begin to serve to establish comparative and baseline data for the prototype new coolants. These tests will include fundamental properties, such as pH value and specific gravity, physical properties, and coolantspecific parameters including foaming tendency and reserve alkalinity. It is believed that this data would



direct the research towards the most appropriate coolants. The results are shown in Table 2.

Table 2: Physical and Chemical Properties

Test Number & Description	Comparative Current Specification Value	Comments
ASTM D-1122 Relative Density An experiment to determine the property of relative density. This information is used later in verifying the quality of commercialised products produced at blending facilities, and also has value to estimate contamination levels.	1.110-1.145	The relative density of the new coolant will be different than EG or PG and will also depend on the concentration of PDO and water.
ASTM D-1177 Freeze Point This experiment overcomes the soft 'slushy' freeze characteristic that makes determining the freezing point of some fluids difficult. It produces a graph of cooling behaviour from which a consistent and meaningful freeze point can be determined.	<-40°C	Choosing an appropriate solution can satisfy this requirement.
ASTM D-1120 Boiling Point This is a boiling point method consistent with standard methods used to determine the boiling points of most fluids.	>90°C	The boiling point of the new coolant will be different than EG or PG and will also depend on the concentration of PDO and
ASTM D-1882 Auto Finish The coolant is likely to be spilled on an auto finish. Therefore, it has always been a requirement that the coolant has no effect on the cars' finish, and this test was developed to evaluate that property.	no effect	water No problem expected.





Test Number & Description	Comparative Current Specification Value	Comments
ASTM D-1119 Ash Content High levels of dissolved solids are associated with premature water pump wear and other durability issues. Completely evaporating the liquid and calculating the weight of the remaining dry material determines ash content.	<5.0% max.	Since this coolant will be very low in inhibitors, this specification may need to be further reduced to prevent conductivity problems.
ASTM D-1287 pH: The H ⁺ ion concentration is reported as a pH value. This value is determined from an instrument reading. The pH value has to be appropriate for the inhibitor technology in use.	7.5 to 11.0	Experimentation will likely result in a tighter spec for PDO than is used today for EG and PG coolants.
ASTM D-1123 Water mass percent Water content on non-aqueous coolants is determined by the Karl Fischer method.	5.0 % max.	Applicable to the PDO before blending.
ASTM D-1121 Reserve Alkalinity In many inhibition technologies, the durability of the coolant is related to its ability to neutralise weak acids formed as the base and/or inhibitors degrade. This titration evaluates that property.		This property may be obsolete, or may have QC value.
ASTM D-1881 Foaming Tendencies Foaming is an undesirable property associated with negative performance. This method creates a measurable volume, and also the time required to dissipate the foam.	Break: 5 sec. Volume: 150 ml	The new coolant should meet this requirement.
Electrical Conductivity mohs Test method: a calibrated laboratory bench conductivity meter is employed to measure the conductivity of the coolant. The conductivity probe is placed into the fluid, and the digital reading on the conductivity meter is observed.	< 50	Experimental data to be used in developing a test and performance specification.
Viscosity mPa.s (cPs) ASTM D-445	<1@80 °C <6@0 °C	Comparable to EG coolant.
Thermal Conductivity W/m-K from literature	>0.4	Comparable to EG coolant.

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The state of the s	Comparative	Comments
Test Number & Description	Current	
	Specification	
	Value	
Heat Capacity (kJ/kg-K) from	>3	Comparable to
literature	}	EG coolant.
V)		1
Durability by extended duration	>5 years	PDO. promises
tests	70012	excellent
		stability.
		- Julian State of the state of
Effect on Elastomers:	<10% Δ	
By Cummins Method 14292	Dimension Each	
Silicon Seals, Viton, Bunan		
(Nitrile), Teflon, Neoprene,		
Rubber,		
Nylon	1	
1,1201		
Toxicity LD ₅₀ data and review of	Non toxic for	PDO offers low
MSDS	transporta-	toxicity.
·	tion.	_
ASTM D2809 Water Pump Test,	≥8 each time	
repeated three times	26 each time	PDO has
repeaced times	1	performed
		better than EG in the series
	1	1
		of tests. See Table 3 below.
ASTM D-4340 Corrosion of Aluminium	<1.0	PDO has
Heat Rejecting Surfaces	mg/cm²/week	performed at
near Rejecting Bullaces	lig/ cli / week	less than 10%
		of the allowed
		loss.
Extended ageing evaluation in D-	<1.0	PDO degraded
4340 Rig @ 150 °C for 60 Days	mg/cm²/week <	less in terms
sampled @ 10 day intervals.	2 pH units	of pH value
Aluminium weight loss	<20%	and in the
Δ pH	< 2,000 ppm	formation of
Oxidation products (i.e. COOH	m<1	oxidation by-
anions)		products in
Oxidation trend (slope of	1	the presence
regression)		of two fully
-	[formulated
	†	coolant
	1	inhibition
		packages. See
	15 - 2	Table 4 below.
ASTM D-1384 Corrosion in Glassware	Maximum Weight	Test passed.
(Higher Performance	Loss, mg	
Specification)	5	
Copper	10	
Lead Solder	5	
Brass	5	
Steel	5	
Cast Iron	10	
Cast Aluminium		
· · · · · · · · · · · · · · · · · · ·	L	Ш

	Comparative	Comments
Test Number & Description	Current	
	Specification	
	Value	
Aged Coolant Corrosion (ASTM D-1384	Maximum Weight	
extended) in Glassware @ 150 °C	Loss, mg	
(Fluid from 2,000 Hour Ageing)	10	
Copper	30	
Lead Solder	10	
Brass	10	
Steel	10	
Cast Iron	30	
Cast Aluminium		
Erosion Corrosion of Heat	No leaks	
Exchanger, 2,000 hours		
Repassivation of Aluminium by	$E_{B} < 2.0$	
Galvanostatic Measurement ASTM	$E_{G} > -0.4.0$	
D6208		_ 1
ASTM D-2570 Simulated Service	Maximum Weight	Multiple
(Higher Performance	Loss, mg	embodiments
Specification)	10	passed.
Copper	20	
Lead Solder	10	
Brass	10	
Steel	10)
Cast Iron	20	
Cast Aluminium		

Table 3

ASTM 2809 Test Data		
Inhibitor	EG	PDO
Conventional Automotive	8	9
Carboxylate Automotive	2	8
Phosphated Heavy Duty	10	10
Non Phosphated Heavy Duty	3	8
Hybrid Heavy Duty	9	10

Table 4

Oxidation comparison between PDO and EG inhibited with commercial inhibitor package @ 2.2%. Test run on D-4340 at 150°C, without corrosive water and at 50% concentration.

			pН				
Time (days)	0	10	20	30	40	50	60
PDO-A	11.16	9.31	8.87	8.69	8.41	8.19	7.96
EG~A	10.06	7.67	6.38	5.68	4.60	4.31	4.07
PDO-B	10.58	9.63	8.89	8.56	8.32	8.18	7.93
EG-B	10.67	9.22	8.67	8.32	8.02	7.92	7.74
		Total	Degrada	tion Ac	ids (ppm	1)	
Time (days)	0	10	20	30	40	50	60
PDO-A	0	213	415	607	762	851	1029
EG-A	0	542	1553	1987	3498	4028	4705
PDO-B	0	231	372	587	688	833	1053
EG-B	0	342	654	922	1128	1486	1602

Example 2

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In these experiments, a solution of 50 percent by volume 1,3-propane diol (PDO) and 50 percent by volume deionised water were tested for corrosion of various metals used in engine cooling systems over a period of time. The test method was modified from ASTM test method D-2570 by using the spaced interval examination procedure detailed in ASTM G-31. The following Table 5 shows the results:

Table 5: Extended Spaced Interval Simulated Service Test Modified from ASTM D2570 (using ASTM G31 spaced interval)
Test Method

PDO @ 50% in DI Water 190 °F (88°C). Spaced Interval Corrosion Data

Weeks	2	4	6	8	10
Copper	2 .	2	1	1	2
Lead Solder	3	2	6	. 6	3
Brass	2	2	3	3	4
Steel	11	12	13	13	13
Cast Iron	13	10	11	11	40
Cast	22	34	40	40	40
Aluminium					

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Note how the corrosion behaves after 8-10 weeks. The fact that the aluminium corrosion does not increase after 6 weeks gives an indication that there is some flash corrosion initially but after that the oxides protect the aluminium. Generally, the absolute limit is specified by ASTM D3306 to be 60mg of aluminium lost after 7 weeks' exposure.

Example 3

The next experiment was corrosion of aluminium services over an extended period of time. The results are set out in Table 6 below.

Table 6: Corrosion of Heat Rejecting Aluminium Surface
Modified from ASTM D4340

Temperature elevated to 149°C (300 $^{\circ}F$), Time extended from 1 week to 30 days 50% PDO 50% (volume) DI Water

	Before Test	10 Days	20 Days	30 Days
Weight loss mg/cm²/week	-	0.0	0.0	0.0
рН	6.55	5.34	4.60	4.99
Conductivity pmhos/cm	0	9	9	14
Comments	No damage to specimen			

Please note that even after running this test for 30 days, there was no apparent corrosion damage to the specimen.

Example 4

This example describes experiments following the ASTM D1384 test method, modified by omitting the corrosive salts and were also made to operate at 150 degrees C by changing the bath from water to 50% propylene glycol. The tests were done to test the corrosivity of solutions of PDO in water

having amounts of PDO from 55 to 85 percent by weight. The 65 weight percent PDO solution was identified as being the best because it offered the best overall protection for the six metals tested. However, the data in Table 7 also shows that solutions containing 55% and 60% PDO in water also achieved very good results because fuel cell systems are most likely to be manufactured primarily of aluminium and stainless steel.

Table 7

					·		
Percent PDO in water	55	60	65	70	75	80	85
Copper	1.2	2.0	1.6	1.7	1.7	1.6	0.6
Lead Solder	123.8	93.5	62.5	60.3	39.2	63.7	20.3
Brass	2.1	1.7	1.8	2.0	1.7	2.7	1.2
Steel	126.1	86.8	84.6	15.8	29.2	26.3	1.5
Cast Iron	247.6	186.6	263	255.3	,227.1	189.3	-0.7
Cast Aluminium	8.2	7.0	7.3	16.6	17.3	47.5	26.5
Conductivity Before	0	0	0	0	0	0	0
Test µmhos/cm							
Conductivity After Test	30	22	10	.7	4	3	0
µmhos/cm			. •				

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Summary of Results

It is believed that the results show that these PDO-based coolants can be used for a low conductivity application in fuel cell powered systems, including fuel cell vehicles. PDO is demonstrated to be non-conductive and manifests corrosion resistant properties to the point of meriting serious consideration. The following are some of the more significant findings:

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• A coolant with high electrical resistance (low conductivity) has been developed that is appropriate for use in fuel cell powered systems, including fuel cell powered vehicles, that generate strong

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electrical fields. It has electrical resistivity of more than 250 kohm-cm. Ethylene glycol is too corrosive to be completely nonconductive.

- The coolant, containing PDO, can be formulated in various concentrations to achieve freeze points of -40 (°F or °C) or lower (see freeze point graphs in Figures 1 and 2).
- The coolant offers more favourable boiling points in aqueous solutions than traditional glycol based coolants, as high as 234°C (471 °F).
- The thermal conductivity is comparable to glycolbased coolants (in water).
- The viscosity is comparable to glycol-based coolants (in water).
- The heat capacity is comparable to glycol-based coolants (in water).
 - The durability is better than glycol based coolants, offering the prospect of a closed, lifetime-filled low or no maintenance coolant system.
- The coolant is compatible with system materials, including aluminium and elastomers.
 - The coolant is less toxic and less palatable than ethylene glycol and is much less likely to be involved in pet or child poisonings.
- The cost of the coolant over the life of the system is comparable to existing premium coolants

The physical property data for PDO and potentially competing coolants, ethylene glycol (EG) and propylene glycol (PG) are shown in Table 8:



Table 8

Physical Properties	PDO	EG	PG
Mol. Wt.	76.1	62.07	76.1
Boiling Point, °C (°F)	214.4	197.6	187.4
·	(417.9)	(387.7)	(369.3)
Flash Point, °C (°F)	129	116	104
	(265)	(240)	(220)
Specific Gravity, 20°C	1.0526	1.115	1.032
Freeze Point, 50% solution, °C(°F)	-29	-38	-33
	(-21)	(-36)	(-28)
Pour Point, °C (°F)		<-59	<-57
		(<-75)	(<-71)
Viscosity, mPa.s (cP) 20°C	52	17	49
Specific Heat, kJ/(kg*K) (212°F	2.730	2.784	2.948
BTU/lb/F)	(0.652)	(0.665)	(0.704)
Thermal Conductivity, W/(m*K) @ 25°C	0.220	0.254	0.206
(25°C BTU/hr-ft-F)	(0.127)	(0.147)	(0.119)
Heat of Vaporisation, kJ/kg @ 25°C	954	1044	882
(25°C BTU/lb)	(410)	(449)	(379)
Purity	99.7	94.5	99

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CLAIMS

- 1. A nontoxic fuel cell engine coolant which has an electrical resistivity of greater than 250 kOhm-cm, a boiling point of greater than 90°C, a thermal conductivity of greater than 0.4 W/m-k, a viscosity of less than 1 mPa.s (1 cPs) at 80°C, a viscosity of less than 6 mPa.s (6 cPs) at 0°C, a heat capacity of greater than 3 kJ/kg-K, and which is compatible with current cooling system materials.
- The coolant of claim 1 wherein the coolant is 1,3 propanediol.
 - 3. The coolant of claim 1 is an aqueous solution comprised of from 1 to 100% by volume of 1,3-propanediol.
 - 4. The coolant of claim 3 wherein the solution is comprised of from 40 to 85% by volume of 1,3-propanediol.
- 5. The coolant of claim 4 wherein the solution is comprised of from 55 to 85% by volume of 1,3-propanediol.
 - 6. The coolant of claim 1 having a freezing point of less than -40°C.

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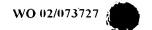


Figure 1

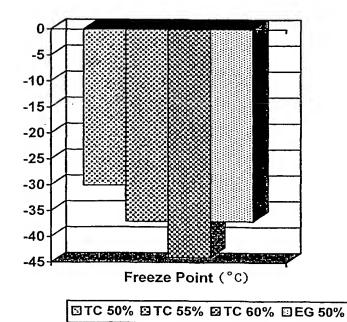
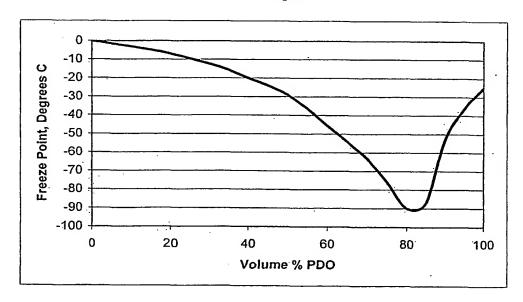


Figure 2



A. CLASSIFI IPC 7	ICATION OF SUBJECT MATTER H01M8/04 C09K5/10		
According to	International Patent Classification (IPC) or to both national class	sification and IPC	
B. FIELDS S			
IPC 7	cumentation searched (classification system followed by classifi CO9K H01M	ication symbols)	
Documentation	on searched other than minimum documentation to the extent th	nat such documents are included in the fields se	arched
	ita base consulted during the international search (name of data ternal, WPI Data, PAJ, CHEM ABS Da	•)
LI O III	icinar, mr baca, mo, chen mo be		
C. DOCUME	NTS CONSIDERED TO BE RELEVANT		
Category °	Citation of document, with indication, where appropriate, of the	e relevant passages	Relevant to claim No.
A	EATON E R ET AL: "A CHEMICAL! ENGINE COOLANT / ANTIFREEZE WI' THERMAL STABILITY PROPERTIES" SAE SPECIAL PUBLICATIONS, XX,	TH IMPROVED	1-6
	2001, pages 55-61, XP008003880 ISSN: 0099-5908 the whole document		
Α	WO 00 33407 A (BALLARD POWER S; WILKINSON DAVID P (CA); ROBER (CA);) 8 June 2000 (2000-06-08 page 15, line 18 -page 16, lin	TS JOY A)	1
	•		
Furth	her documents are listed in the continuation of box C.	γ Patent family members are listed	in annex.
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consid	ent defining the general state of the art which is not lered to be of particular relevance	"T" later document published after the into or priority date and not in conflict with cited to understand the principle or the invention	the application but
filing d	document but published on or after the international late ent which may throw doubts on priority claim(s) or is cited to establish the publication date of another	*X* document of particular relevance; the cannot be considered novel or cannot involve an inventive step when the	ot be considered to ocument is taken alone
citation 'O' docume other	n or other special reason (as specified) ent referring to an oral disclosure, use, exhibilion or means	"Y" document of particular relevance; the cannot be considered to involve an in document is combined with one or ments, such combination being obvious.	oventive step when the ore other such docu-
later ti	ent published prior to the international filing date but han the priority date claimed	in the art. *&* document member of the same paten	
	actual completion of the international search	Date of mailing of the international se	earch report
	B July 2002	16/07/2002 Authorized officer	
.vame and	European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epo nl, Fay: (431-70) 340-3016	Puetz. C	

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intormation on patent family members

T/EP 02/01801

*				1/ 21	02/01001
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WO 0033407	A	08-06-2000	AU WO GB US	1369200 A 0033407 A1 2370407 A 2002006534 A1	19-06-2000 08-06-2000 26-06-2002 17-01-2002

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